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Assessment of $Pr^{3+}:KY_3F_{10}$ as a Blue-Green Laser

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The reported energy levels of praseodymium and europium in the host material KY_3F_{10} have been analyzed. Using the x-ray data, lattice sums have been done, and odd crystal-field parameters have been calculated. Using these results, the transition probabilities for both magnetic and electric dipoles have been calculated for the rare earth ions. Discussions are given of the application of the results to a blue-green laser using praseodymium.

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CONTENTS

TABLES

1. INTRODUCTION

The most promising blue-green laser crystal reported[1] to date is Pr^{3+}:YLF (praseodymium in lithium yttrium fluoride, $LiYF_4$ or YLF). The only strong transitions from the $^3P_0$ fluorescing level to the $^3H_4$ ground state multiplet of Pr^{3+} are, unfortunately, to the lowest lying levels of $^3H_4$, which means that the laser is a three-level system even at low temperature (77 K). Since four-level laser action is necessary to achieve low lasing threshold and high efficiency, other hosts where the laser terminates on an upper energy level are desired, providing that the material has other required properties for blue-green laser operation such as a large band gap and good optical quality. One possible material is Pr^{3+}:KY_3F_{10}, which was examined in this work as described below.

X-ray data[2] were used in a lattice sum calculation, as is discussed in section 2, to determine the spherical tensor amplitudes, A_{kq}, of the electrostatic field at the impurity ion site. By using these A_{kq} in a three-parameter model[3] of crystal fields, it was possible to estimate the energy levels of europium, Eu^{3+}, in KY_3F_{10}, which were obtained from the optical data recorded[4] by Porcher and Caro for Eu^{3+}:KY_3F_{10} powder. These data were then successfully analyzed, and phenomenological crystal field parameters, B_{kq}, were found by a method described elsewhere[5] that gave a least-squares fit of 6.9 cm^{-1} between 26 calculated and measured energy levels for Eu^{3+}.

The above Eu^{3+} parameters were scaled[6] by the appropriate ρ_k values, $B_{kq} = \rho_k$ (ion dependent) A_{kq} (host dependent), to give even-fold (even-k) B_{kq} required to predict the Stark split energy levels for neodyminum, ND^{3+}, and Pr^{3+}; the method of calculation is described elsewhere.[5] One of the authors had available preliminary (and unpublished) energy levels and optical data for both Nd^{3+}:KY_3F_{10} and PR^{3+}:KY_3F_{10}. These data were analyzed, and phenomenological B_{kq} were found that gave root mean square (rms) deviations of 6.3 cm^{-1} and 14.3 cm^{-1}, respectively, between 23 Nd^{3+} and 35 Pr^{3+} calculated and measured energy levels.

By using the even-k phenomenological B_{kq} for Pr^{3+} and the odd-k A_{kq} obtained from the lattice sum, electric and magnetic dipole transition probabilities and branching ratios were calculated. According to the branching ratio calculations, about 33 percent of the electromagnetic radiation from the $^3P_0$ level goes to the ground multiplet, which compares[7] with 36 percent going to the ground multiplet for Pr^{3+} in YLF. The largest transition goes to the lower energy states, as for Pr^{3+} in YLF. However, two of the largest three transitions from $^3P_0$ in KY_3F_{10} go to levels at 166 and 217 cm^{-1}, whereas one of the larger transitions goes to a level at 79 cm^{-1} in YLF. Thus, four-level operation of Pr^{3+} in KY_3F_{10} at 77 K might be attained since the possible terminal level would not be so thermally populated as it is in YLF.

[1] L. Esterowitz, R. Allen, M. Kruer, F. Bartoli, L. S. Goldberg, H. P. Jenssen, A. Linz, and V. Nicolai, J. Appl. Phys., 48 (1977), 650.

[2] J. W. Pierce and H. Y.-P. Hong, Proceedings of Tenth Rare-Earth Research Conference (1973), 527.

[3] Richard P. Leavitt, Clyde A. Morrison, and Donald E. Wortman, Rare-Earth Ion-Host Crystal Interactions 3. Three-Parameter Theory of Crystal Fields, Harry Diamond Laboratories HDL-TR-1673 (June 1975).

[4] P. Porcher and P. Caro, J. Chem. Phys., 65 (1976), 89.

[5] N. Karayianis, D. E. Wortman, and H. P. Jenssen, J. Phys. Chem. Solids, 37 (1976), 675.

[6] Clyde A. Morrison, Nick Karayianis, and Donald E. Wortman, Rare-Earth Ion-Host Lattice Interactions 4. Predicting Spectra and Intensities of Lanthanides in Crystals, Harry Diamond Laboratories HDL-TR-1816 (June 1977).

[7] Leon Esterowitz, Filbert J. Bartoli, Roger E. Allen, Donald E. Wortman, Clyde A. Morrison, and Richard P. Leavitt, Energy Levels and Line Intensities for the Ground Configuration of Pr^{3+} in $LiYF_4$, Harry Diamond Laboratories HDL-TR-1875 (February 1979).

In addition to the above calculations, phenomenological A_{kq} were determined by the relation $B_{kq} = \rho_k A_{kq}$ and from the phenomenological B_{kq} determined in the Pr^{3+}, Nd^{3+}, and Eu^{3+} spectral analyses. These A_{kq}, which should depend only upon the host crystal, were then averaged to give a set of A_{kq} at a Y site. This set of A_{kq} were multiplied by appropriate ρ_k for each lanthanide ion, and B_{kq} were obtained for the entire lanthanide series. Energy levels for each of the triply ionized lanthanides in KY_3F_{10} were calculated by using the appropriate set of B_{kq}, and these results are given.

2. CRYSTALLOGRAPHIC DATA AND LATTICE SUM FOR KY_3F_{10}

The first complete x-ray analysis of KY_3F_{10} was done by Pierce and Hong.[2] The space group most consistent with their x-ray data is Fm3m (space group 225 of the international tables[8]) with all the Y ions at sites of C_{4v} symmetry. A second possible space group, F$\bar{4}$3m, fit Pierce and Hong's data as well as the Fm3m, but the Fm3m was chosen by them because it is more symmetric than F$\bar{4}$3m. This choice has been confirmed by recent optical data[4] on Eu^{3+} in KY_3F_{10}. If the space group were F$\bar{4}$3m, the Eu^{3+} ion would occupy a site of C_{2v} symmetry, and complete removal of the degeneracy of the free ion would be observed. However, the optical spectra agree with what should be observed if the ion were in a site of C_{4v} symmetry, which agrees with the symmetry of the Y site of the space group Fm3m.

Recently, the material potassium ytterbium fluoride (KYb_3F_{10}) has been grown[9] and found to have the same space group, Fm3m, with the cubic cell dimension, a = 11.431 Å, and the positions of the constituent ions are $x_{Yb} = 0.24$, $x_{F_1} = 0.33$, and $x_{F_2} = 0.12$. A large number of compounds also have been grown recently by Vedrine et al,[10] which are cubic and have the form KLn_3F_{10} (Ln = lutetium—Lu, Yb, thulium—Tm, erbium—Er, holmium—Ho, or dysprosium—Dy) and $RbLn_3F_{10}$ (Ln = terbium—Tb, gadolinium—Gd, Eu, or samarium—Sm) with only the cubic cell dimension reported.

We have performed the lattice sum for KY_3F_{10} with the origin at a Y site using Pierce and Hong's[2] data as given in table 1 with the various ions occupying the sites given in the international tables,[8] and the results are given in table 2. There are 24 Y ions in a unit cell, and each site has C_{4v} symmetry. However, eight Y ions at (0, 0, ±x) have the principal axis of C_{4v} along z, eight Y ions at (±x, 0, 0) have the principal axis along x, and eight Y ions at (0, ±x, 0) have the principal axis along y. The full 24 Y ions are generated by three additional translations within the unit cell: (0, ½, ½), (½, 0, ½), and (½, ½, 0), which preserve the axis of symmetry of each particular site. Whereas the optical spectra of a rare-earth ion at a single site possessing C_{4v} symmetry have well-defined polarization selection rules, the multiplicity of the principal axes causes polarization data to be meaningless. Because of the multiplicity of the principal axes of C_{4v} symmetry in KY_3F_{10}, a magnetic field applied in an arbitrary direction gives rise to three sets of Zeeman splittings in the spectrum of a rare-earth ion. These sets of lines interchange as the direction of the magnetic field is varied. It is possible that the only advantage of using a magnetic field in analyzing the optical spectra would be for those rare-earth ions possessing an even number of electrons. The magnetic field could then be used to readily identify the doublets existing for these ions in C_{4v} symmetry.

[2] J. W. Pierce and H. Y.-P. Hong, Proceedings of Tenth Rare-Earth Research Conference (1973), 527.
[4] P. Porcher and P. Caro, J. Chem. Phys., *65* (1976), 89.
[8] International Tables, *I*, Kynoch Press, Birmingham, U.K. (1952), 338, table 225.
[9] M. Labean, S. Aleonard, A. Vedrine, R. Boutonnet, and J. C. Cousseins, Mat. Res. Bull., *9* (1974), 615.
[10] A. Vedrine, R. Boutonnet, R. Sabatier, and J. Cousseins, Bull. Soc. Chim. Fr., *3-4* (1975), 445.

TABLE 1. CRYSTAL STRUCTURE DATA ON KY_3F_{10}
Space Group Fm3m cubic a = 11.542 ± 0.004 Å

Ion	Position	Site symmetry	Ion[a] position
Y	24e	C_{4v}	x = 0.2401
K	8c	T_d	—
F_1	48i	C_{2v}	x = 0.3353
F_2	32f	C_{3v}	x = 0.1081

[a] These are the ion positions that have to be determined by x-ray analysis for use in the International Tables. The position 8c is determined in terms of rational fractions.

Sources: J. W. Pierce and H. Y.-P. Hong, Proceedings of Tenth Rare-Earth Research Conference (1973), 527; International Tables, 1, Kynoch Press, Birmingham, U.K. (1952), 338, table 225.

TABLE 2. LATTICE SUM FOR KY_3F_{10}

Spherical tensor amplitudes	
A_{kq}	$(cm^{-1}/(Å)^k)$
A_{10}	4749.97
A_{20}	−4011.53
A_{30}	−4431.00
A_{40}	−4822.27
A_{44}	1531.83
A_{50}	1048.16
A_{54}	2799.43
A_{60}	661.89
A_{64}	42.32
A_{70}	54.69
A_{74}	241.86

Note: The origin is the yttrium ion at (0, 0, ±x). The effective charges are the normal valence values.

Source of x-ray data: J. W. Pierce and H. Y.-P. Hong, Proceedings of Tenth Rare-Earth Research Conference (1973), 527.

3. ANALYSIS OF Eu^{3+}, Nd^{3+}, AND Pr^{3+} OPTICAL DATA

In making the energy level calculations, the crystal field Hamiltonian,

$$H_x = \sum_{kq} B_{kq} C_{kq} , \qquad (1)$$

where C_{kq} are spherical tensors, was diagonalized in a basis of states spanning a particular set of free-ion J-multiplets by the method described elsewhere.[5] The B_{kq} are related to the A_{kq}, which were obtained in the lattice sum, by the expression,

$$B_{kq} = \rho_k A_{kq} . \qquad (2)$$

The ρ_k were obtained[3,6] by considering shielding and radial wave-function expansion and are ion-dependent quantities given by Morrison et al in their table II.[6]

[3] Richard P. Leavitt, Clyde A. Morrison, and Donald E. Wortman, Rare-Earth Ion-Host Crystal Interactions 3. Three-Parameter Theory of Crystal Fields, Harry Diamond Laboratories HDL-TR-1673 (June 1975).

[5] N. Karayianis, D. E. Wortman, and H. P. Jenssen, J. Phys. Chem. Solids, 37 (1976), 675.

[6] Clyde A. Morrison, Nick Karayianis, and Donald E. Wortman, Rare-Earth Ion-Host Lattice Interactions 4. Predicting Spectra and Intensities of Lanthanides in Crystals, Harry Diamond Laboratories HDL-TR-1816 (June 1977).

In this work, the A_{kq} given in table 2 for KY_3F_{10} were multiplied by the ρ_k values[6] for Eu^{3+}. This procedure gave a set of B_{kq}, which were used to calculate an initial set of energy levels for $Eu^{3+}:KY_3F_{10}$. These calculated levels were compared with the experimental values of Porcher and Caro[4] in a least-squares fit calculation. The B_{kq} were varied, and new calculated energy levels were determined in the fitting procedure until a least rms value of 6.9 cm^{-1} between 26 calculated and experimental levels was obtained. The phenomenological B_{kq} yielding this fit for Eu^{3+} are given in table 3. Porcher and Caro[4] reported B_{kq} for Eu^{3+}, and these values are given for comparison in table 3.

TABLE 3. PHENOMENOLOGICAL CRYSTAL FIELD PARAMETERS (B_{kq}) FOR TRIPLY IONIZED LANTHANIDES IN KY_3F_{10}

Parameter	Pr^a	Nd^a	Eu^b	Eu^c
B_{20} (cm^{-1})	−589.0	−621.1	−517.6	−551
B_{40} (cm^{-1})	−1711	−1566	−1337	−1360
B_{44} (cm^{-1})	518.9	252.2	359.6	345
B_{60} (cm^{-1})	656.9	366.7	400.1	394
B_{64} (cm^{-1})	−195.9	−366.6	−2[illegible]	−234
Root mean square	14.3	6.3	6.9	7.6
Levels in fit (No.)	35	23	26	25
Multiplets with data (No.)	12	5	8	8
Crystal field splittings (No.)	23	18	18	16

[a] Preliminary data from H. P. Jenssen, Massachusetts Institute of Technology (unpublished).
[b] Our fit on data reported by P. Porcher and P. Caro, J. Chem. Phys., 65 (1976), 89.
[c] B_{kq} reported by Porcher and Caro. B_{64} should be −234 rather than +234 (private communication).

The phenomenological B_{kq} for Eu^{3+} that we obtained were scaled by the ratio $\rho_k(Nd)/\rho_k(Eu)$ to give an initial set of B_{kq} for Nd^{3+}. This initial set of B_{kq} was used to calculate energy levels for Nd^{3+} in KY_3F_{10} for comparison with our preliminary data. Phenomenological B_{kq} for Nd^{3+} were obtained that yielded a least rms value of 6.3 cm^{-1} between 23 calculated and measured levels; these B_{kq} are given in table 3.

Initial B_{kq} were obtained for Pr^{3+} in KY_3F_{10} by using the phenomenological B_{kq} for Eu^{3+}. These B_{kq} were used to calculate energy levels that aided us in identifying 35 energy levels in the Pr^{3+} ground configuration. Phenomenological B_{kq} listed in table 3, gave a least rms deviation of 14.3 cm^{-1} between 35 calculated and measured Pr^{3+} energy levels.

4. CALCULATIONS AND DISCUSSION OF RESULTS

The above phenomenological B_{kq} for Pr, Nd, and Eu were divided by the appropriate[6] ρ_k values, and the phenomenological A_{kq} values listed in table 4 were obtained. Since the A_{kq} are supposed to be host dependent only, the variations in the A_{kq} give some idea about the uncertainty in these parameters. The even-k A_{kq} determined from these three ions were averaged to give a set of A_{kq} for KY_3F_{10}; these values are given in table 4 along with the A_{kq} obtained in the lattice sums for two different charges on the F ions. In addition, the averaged A_{kq} were multiplied by the appropriate[6] ρ_k, and B_{kq} were determined for the lanthanides in KY_3F_{10} as given in table 5. Energy levels calculated by using the B_{kq} given in table 5 for each of the lanthanides in KY_3F_{10} are given in tables 6 to 18. In the tables, 2 MU is twice the crystal quantum number,[11] 2μ.

[4] P. Porcher and P. Caro, J. Chem. Phys., 65 (1976), 89.

[6] Clyde A. Morrison, Nick Karayianis, and Donald E. Wortman, Rare-Earth Ion-Host Lattice Interactions 4. Predicting Spectra and Intensities of Lanthanides in Crystals, Harry Diamond Laboratories HDL-TR-1816 (June 1977).

[11] K. H. Hellwege, Ann. Physik, 4 (1948), 95.

TABLE 4. EVEN-K PHENOMENOLOGICAL A_{kn} AND A_{kn} DETERMINED FROM SUMMATION OVER KY_3F_{10} LATTICE

Amplitude or radial factor	Calculated crystal field parameter					
	1	2	3	4	5	6
A_{20}	−3354	−3641	−3107	−3367	−4011	−4288
A_{40}	−2647	−2711	−2765	−2708	−4822	−4752
A_{44}	802.7	436.6	743.6	661.0	1532	1493
A_{60}	350.3	230.7	320.0	300.3	661.9	654.7
A_{64}	−104.5	−230.6	−217.3	−184.1	42.32	42.1
ρ_2	0.1756	0.1706	0.1666	—	—	—
ρ_4	0.6464	0.5776	0.4836	—	—	—
ρ_6	1.8754	1.5897	1.2503	—	—	—

Notes:

1: $A_{kn} = B_{kn}/\rho_k$; Pr^{3+} B_{kn} and ρ_k.
2: $A_{kn} = B_{kn}/\rho_k$; Nd^{3+} B_{kn} and ρ_k.
3: $A_{kn} = B_{kn}/\rho_k$; Eu^{3+} B_{kn} and ρ_k.
4: Average of 1, 2, and 3.
5: A_{kn} from lattice sum where the charges used in the sum are $q_K = 1$, $q_Y = 3$, and $q_F = -1$.
6: A_{kn} from lattice sum with $q_K = 0.9$, $q_Y = 3$, and $q_F = -0.99$.

TABLE 5. CRYSTAL FIELD PARAMETERS FOR TRIPLY IONIZED LANTHANIDES IN KY_3F_{10}

Ion	N^a	N^b	B_{20}	B_{40}	B_{44}	B_{60}	B_{64}	Root mean square deviation
Ce	0	7	−620	−2041	498	703	−431	—
Pr	35	70	−589	−1711	519	657	−196	14.3
Nd	23	60	−621	−1566	252	367	−367	6.3
Pm	0	89	−565	−1446	353	427	−262	—
Sm	0	59	−562	−1367	334	397	−243	—
Eu	26	59	−518	−1337	360	400	−272	6.9
Gd	0	67	−562	−1261	308	356	−219	—
Tb	0	75	−563	−1216	297	337	−207	—
Dy	0	67	−566	−1176	287	319	−195	—
Ho	0	68	−570	−1142	279	304	−186	—
Er	0	59	−574	−1117	273	295	−181	—
Tm	0	70	−580	−1098	268	290	−178	—
Yb	0	7	−595	−1056	260	274	−168	—

[a] Number of experimental levels.
[b] Number of theoretical levels.
Note: Units are in cm^{-1}.

TABLE 6. ENERGY LEVELS FOR Ce^{3+} IN KY_3F_{10}

LSJ state (centroid)		Energy level			
$^2F_{5/2}$	2 MU	1	3	3	
(250)	TH.	0	342	600	
$^2F_{7/2}$	2 MU	1	3	1	3
(2550)	TH.	2271	2624	2695	3074

TABLE 7. ENERGY LEVELS FOR Pr^{3+} IN KY_3F_{10}

LSJ state (centroid)		Energy level							
3H 4	2 MU	2	4	0	4	2	0	0	
(232)	TH.	−7	69	161	183	223	511	541	
	EXP.	0		166	175	217			
3H 5	2 MU	2	0	4	0	2	4	0	2
(2361)	TH.	2197	2222	2226	2280	2313	2334	2393	2656
	EXP.	2184		2211					2684
3H 6 +	2 MU	0	2	0	4	2	0	4	2
3F 2	TH.	4290	4354	4371	4430	4452	4524	4531	4562
(4546,	EXP.	4283	4347		4418	4477			
5080)	2 MU	4	4	4	4	2	0		
	TH.	4991	4993	5031	5071	5100	5214		
	EXP.			5041		5075	5229		
3F 3	2 MU	2	4	4	2	0			
(6466)	TH.	6422	6450	6458	6499	6573			
	EXP.	6433	6441		6496	6573			
3F 4	2 MU	2	4	4	0	0	2	0	
(6928)	TH.	6801	6804	6909	6994	7006	7068	7150	
	EXP.	6829		6895		7006	7055		
1G 4	2 MU	4	2	4	2	0	0	0	
(9945)	TH.	9622	9750	9845	10064	10080	10085	10233	
	EXP.	9622							
1D 2	2 MU	4	0	4	2				
(16967)	TH.	16669	16820	16907	17205				
	EXP.	16654		16890	17237				
3P 0 +	2 MU	0	0	2	4	4	0	2	4
1I 6 +	TH.	20732	20772	20825	20943	21135	21204	21333	21335
3P 1	EXP.	20728	20794				21204		
(20739,	2 MU	4	2	0	0	2			
21260,	TH.	21342	21358	21613	21635	21709			
21307)	EXP.		21358			21692			
3P 2	2 MU	4	4	0	2				
(22467)	TH.	22343	22376	22570	22591				
	EXP.	22355	22380	22557	22587				
1S 0	2 MU	0							
(46900)	TH.	46911							

TABLE 8. ENERGY LEVELS FOR Nd^{3+} IN KY_3F_{10}

LSJ state (centroid)		Energy level							
4I $\frac{9}{2}$	2 MU	3	1	3	1	1			
(161)	TH.	−11	120	128	195	323			
	EXP.	0	120	131	182	320			
4I $\frac{11}{2}$	2 MU	1	3	1	3	1	3		
(2066)	TH.	1987	1991	2048	2052	2094	2203		
	EXP.	1979	1986	2049	2053	2104			
4I $\frac{13}{2}$	2 MU	1	1	3	3	3	1	3	
(4032)	TH.	3935	3958	3959	4025	4065	4076	4193	
	EXP.	3928	3967	3956		4063	4078	4193	
4I $\frac{15}{2}$	2 MU	1	1	3	3	3	1	3	1
(6050)	TH.	5894	5903	5935	5983	6116	6157	6172	6248
	EXP.	5886			5988		6165	6165	6250
4F $\frac{3}{2}$	2 MU	3	1						
(11512)	TH.	11454	11543						
	EXP.	11450	11547						
4F $\frac{5}{2}$ +	2 MU	3	1	1	3	1	1	3	3
2H $\frac{9}{2}$	TH.	12394	12475	12523	12524	12543	12621	12624	12673
(12480, 12590)									
4F $\frac{7}{2}$ +	2 MU	1	3	1	3	3	1		
4S $\frac{3}{2}$	TH.	13405	13491	13494	13513	13533	13565		
(13500, 13500)									
4F $\frac{9}{2}$	2 MU	1	3	1	3	1			
(14700)	TH.	14601	14663	14690	14758	14817			
2H $\frac{11}{2}$	2 MU	3	1	3	1	3	1		
(15870)	TH.	15851	15863	15863	15871	15890	15900		
4G $\frac{5}{2}$ +	2 MU	3	1	3	1	1	3	3	
2G $\frac{7}{2}$	TH.	17199	17209	17365	17463	17483	17502	17605	
(17300, 17460)									

TABLE 9. ENERGY LEVELS FOR Pm^{3+} IN KY_3F_{10}

LSJ state (centroid)		Energy level							
5I 4	2 MU	0	0	0	2	4	2	4	
(150)	TH.	0	11	66	109	179	185	300	
5I 5	2 MU	2	0	4	2	4	0	0	2
(1577)	TH.	1449	1553	1565	1567	1594	1594	1608	1612
5I 6	2 MU	4	4	2	0	4	2	4	2
(3186)	TH.	3052	3053	3159	3167	3173	3193	3202	3210
	2 MU	0	0						
	TH.	3213	3223						
5I 7	2 MU	2	0	2	4	4	4	4	2
(4850)	TH.	4703	4801	4806	4807	4814	4821	4883	4886
	2 MU	2	0	0					
	TH.	4896	4904	4910					
5I 8	2 MU	0	0	0	2	2	4	4	4
(6600)	TH.	6433	6434	6475	6479	6503	6510	6622	6633
	2 MU	4	2	2	0	0			
	TH.	6668	6694	6711	6721	6731			
5F 1	2 MU	0	2						
(12400)	TH.	12327	12398						
5F 2	2 MU	2	4	0	4				
(12820)	TH.	12729	12806	12882	12887				
5F 3	2 MU	4	2	4	2	0			
(13600)	TH.	13490	13556	13557	13652	13661			
5S 2	2 MU	4	2	4	0				
(14300)	TH.	14285	14286	14287	14291				
5F 4	2 MU	0	2	4	2	4	0	0	
(14650)	TH.	14606	14622	14629	14638	14664	14666	14673	
5F 5 +	2 MU	0	2	4	0	2	0	2	4
3K 6	TH.	15762	15795	15854	15870	15872	15872	15875	15878
(15900,	2 MU	4	2	0	2	4	4	4	2
15900)	TH.	15884	15884	15889	15894	15913	15924	15928	15979
	2 MU	0	0						
	TH.	16006	16009						

TABLE 10. ENERGY LEVELS FOR Sm^{3+} IN KY_3F_{10}

LSJ state (centroid)		Energy level							
6H $\frac{5}{2}$	2 MU	3	1	3					
(36)	TH.	0	219	273					
6H $\frac{7}{2}$	2 MU	1	3	3	1				
(1080)	TH.	1107	1186	1297	1299				
6H $\frac{9}{2}$	2 MU	1	1	3	3	1			
(2286)	TH.	2320	2363	2455	2500	2505			
6H $\frac{11}{2}$	2 MU	3	1	1	3	1	3		
(3608)	TH.	3607	3688	3771	3796	3807	3831		
6H $\frac{13}{2}$	2 MU	3	3	1	3	1	1	3	
(5000)	TH.	4919	5109	5165	5172	5189	5218	5220	
6H $\frac{15}{2}$ +	2 MU	1	1	1	3	3	3	1	3
6F $\frac{1}{2}$ +	TH.	6289	6559	6633	6638	6659	6719	6734	6762
6F $\frac{3}{2}$	2 MU	1	3	1					
(6508,	TH.	6765	6794	6809					
6400,									
6630)									
6F $\frac{5}{2}$	2 MU	3	3	1					
(7100)	TH.	7245	7275	7296					
6F $\frac{7}{2}$	2 MU	3	3	1	1				
(8000)	TH.	8131	8157	8189	8199				
6F $\frac{9}{2}$	2 MU	1	1	3	3	1			
(9200)	TH.	9339	9351	9365	9367	9411			
6F $\frac{11}{2}$	2 MU	1	3	3	3	1	1		
(10500)	TH.	10607	10623	10664	10679	10701	10723		
4G $\frac{5}{2}$	2 MU	3	3	1					
(17900)	TH.	17907	18012	18223					
4F $\frac{3}{2}$	2 MU	3	1						
(18900)	TH.	19045	19071						

TABLE 11. ENERGY LEVELS FOR Eu^{3+} IN KY_3F_{10}

LSJ state (centroid)		Energy level							
7F 0	2 MU	0							
(27)	TH.	0							
	EXP.	0							
7F 1	2 MU	0	2						
(392)	TH.	281	408						
	EXP.	278	411						
7F 2	2 MU	2	4	0	4				
(1067)	TH.	934	1041	1142	1154				
	EXP.	933	1030	1148	1159				
7F 3	2 MU	0	2	2	4	4			
(1927)	TH.	1858	1890	1914	1988	2019			
	EXP.	1858	1895	1903	2002	2012			
7F 4	2 MU	0	0	0	2	4	2	4	
(2894)	TH.	2754	2775	2792	2838	2925	3020	3059	
	EXP.	2748	2778	2800	2845		3014	3052	
7F 5	2 MU	2	4	0	2	4	2	0	0
(3941)	TH.	3742	3926	3931	3940	3975	4005	4021	4064
	EXP.	3739	3929						
7F 6	2 MU	2	0	0	4	4	2	4	4
(4980)	TH.	4822	4876	4884	4936	4938	5008	5096	5138
	2 MU	2	0						
	TH.	5181	5200						
5D 0	2 MU	0							
(17277)	TH.	17276							
5D 1	2 MU	0	2						
(19031)	TH.	19001	19045						
	EXP.	19007	19039						
5D 2	2 MU	0	4	4	2				
(21500)	TH.	21464	21488	21512	21517				
	EXP.	21458	21478	21514	21530				
5D 3	2 MU	4	2	4	2	0			
(24408)	TH.	24386	24392	24404	24420	24444			
5L 6	2 MU	4	4	0	2	4	4	2	2
(25400)	TH.	25204	25209	25242	25266	25333	25414	25486	25566
	2 MU	0	0						
	TH.	25576	25591						

TABLE 12. ENERGY LEVELS FOR Gd^{3+} IN KY_3F_{10}

LSJ state (centroid)		Energy level							
8S $\frac{7}{2}$	2 MU	1	3	3	1				
(0)	TH.	0	0	0	1				
6P $\frac{7}{2}$	2 MU	1	3	3	1				
(32200)	TH.	32147	32172	32213	32252				
6P $\frac{5}{2}$	2 MU	1	3	3					
(32780)	TH.	32743	32775	32806					
6P $\frac{3}{2}$	2 MU	3	1						
(33350)	TH.	33331	33364						
6I $\frac{7}{2}$	2 MU	1	3	1	3				
(35930)	TH.	35907	35929	35931	35957				
6I $\frac{9}{2}$ +	2 MU	1	3	1	3	1	3	1	1
6I $\frac{17}{2}$	TH.	36241	36257	36269	36285	36300	36339	36339	36339
(36270,	2 MU	3	3	1	1	3	1		
36340)	TH.	36339	36340	36340	36342	36342	36344		
6I $\frac{11}{2}$ +	2 MU	1	3	3	3	1	1	1	3
6I $\frac{13}{2}$ +	TH.	36527	36540	36554	36564	36584	36590	36630	36637
6I $\frac{15}{2}$	2 MU	3	3	1	1	3	1	3	1
(36560,	TH.	36648	36657	36673	36687	36687	36690	36693	36696
36660,	2 MU	3	1	3	1	3			
36710)	TH.	36705	36719	36732	36733	36739			
6D $\frac{9}{2}$	2 MU	3	1	3	1	1			
(39720)	TH.	39668	39675	39710	39729	39816			
6D $\frac{1}{2}$	2 MU	1							
(40560)	TH.	40550							
6D $\frac{7}{2}$	2 MU	1	3	1	3				
(40700)	TH.	40689	40699	40701	40707				
6D $\frac{3}{2}$	2 MU	1	3						
(40850)	TH.	40836	40877						
6D $\frac{5}{2}$	2 MU	1	3	3					
(41000)	TH.	40978	40994	41057					

TABLE 13. ENERGY LEVELS FOR Tb^{3+} IN KY_3F_{10}

LSJ state (centroid)		Energy level							
7F 6	2 MU	0	2	4	4	2	4	4	0
(85)	TH.	0	22	71	114	203	288	288	319
	2 MU	0	2						
	TH.	324	384						
7F 5	2 MU	0	0	2	4	2	4	0	2
(2100)	TH.	2128	2159	2163	2186	2211	2219	2226	2415
7F 4	2 MU	4	2	4	2	0	0	0	
(3356)	TH.	3342	3373	3448	3526	3575	3591	3602	
7F 3	2 MU	4	4	2	2	0			
(4400)	TH.	4432	4461	4535	4552	4595			
7F 2	2 MU	4	0	4	2				
(5038)	TH.	5093	5101	5193	5290				
7F 1	2 MU	2	0						
(5440)	TH.	5560	5688						
7F 0	2 MU	0							
(5700)	TH.	5865							
5D 4	2 MU	4	2	4	2	0	0	0	
(20500)	TH.	20580	20598	20607	20624	20634	20692	20692	
5D 3 +	2 MU	2	0	4	4	2	0	2	4
5G 6	TH.	26434	26452	26452	26465	26467	26495	26524	26580
(26336,	2 MU	4	2	0	0	2	4	4	
26500)	TH.	26601	26665	26672	26673	26678	26744	26745	
5L 10	2 MU	2	0	0	4	4	2	0	2
(27100)	TH.	27042	27069	27074	27076	27078	27129	27205	27216
	2 MU	0	2	4	4	2	0	4	4
	TH.	27223	27271	27298	27373	27383	27391	27502	27502

TABLE 14. ENERGY LEVELS FOR Dy^{3+} IN KY_3F_{10}

LSJ state (centroid)		Energy level							
6H $\frac{15}{2}$	2 MU	1	3	1	3	3	1	3	1
(52)	TH.	0	7	31	42	95	97	147	460
6H $\frac{13}{2}$	2 MU	3	1	3	1	1	3	3	
(3517)	TH.	3480	3502	3530	3537	3554	3633	3781	
6H $\frac{11}{2}$	2 MU	1	3	3	1	1	3		
(5850)	TH.	5811	5828	5862	5925	5991	6023		
6H $\frac{9}{2}$ +	2 MU	3	1	1	3	1	3	3	1
6F $\frac{11}{2}$	TH.	7638	7647	7677	7698	7713	7758	7792	7832
(7700,	2 MU	3	1	1					
7700)	TH.	7864	7875	7896					
6H $\frac{7}{2}$ +	2 MU	3	1	1	1	3	3	1	3
6F $\frac{9}{2}$	TH.	9021	9057	9114	9178	9180	9193	9208	9250
(9100,	2 MU	1							
9100)	TH.	9304							
6H $\frac{5}{2}$	2 MU	3	1	3					
(10200)	TH.	10163	10195	10435					
6F $\frac{7}{2}$	2 MU	1	1	3	3				
(11000)	TH.	11042	11057	11087	11133				
6F $\frac{5}{2}$	2 MU	1	3	3					
(12400)	TH.	12445	12465	12514					
6F $\frac{3}{2}$	2 MU	1	3						
(13250)	TH.	13319	13336						
6F $\frac{1}{2}$	2 MU	1							
(13760)	TH.	13839							
4F $\frac{9}{2}$	2 MU	3	1	3	1	1			
(21100)	TH.	21035	21071	21179	21194	21280			
4I $\frac{15}{2}$	2 MU	1	1	3	3	3	1	3	1
(22100)	TH.	22062	22076	22101	22112	22201	22214	22232	22377

TABLE 15. ENERGY LEVELS FOR Ho^{3+} IN KY_3F_{10}

LSJ state (centroid)		Energy level							
5I 8	2 MU	0	0	2	2	4	4	4	4
(108)	TH.	0	8	18	30	52	82	88	156
	2 MU	2	0	2	0	0			
	TH.	167	182	190	246	256			
5I 7	2 MU	0	0	2	2	4	4	2	0
(5130)	TH.	5075	5080	5084	5094	5094	5135	5144	5149
	2 MU	4	4	2					
	TH.	5154	5154	5251					
5I 6	2 MU	0	0	2	4	2	4	0	2
(8580)	TH.	8538	8545	8547	8553	8564	8573	8580	8586
	2 MU	4	4						
	TH.	8692	8692						
5I 5	2 MU	2	4	0	4	2	0	0	2
(11120)	TH.	11087	11092	11099	11110	11111	11116	11130	11215
5I 4	2 MU	4	4	2	2	0	0	0	
(13300)	TH.	13175	13258	13261	13310	13333	13409	13417	
5F 5	2 MU	0	0	2	4	2	4	2	0
(15500)	TH.	15415	15418	15441	15492	15523	15533	15580	15601
5F 4 +	2 MU	0	0	4	4	0	2	2	2
5S 2	TH.	18434	18439	18477	18481	18481	18507	18527	18534
(18500,	2 MU	4	4	0					
18500)	TH.	18538	18545	18558					

TABLE 16. ENERGY LEVELS FOR Er^{3+} IN KY_3F_{10}

LSJ state (centroid)		Energy level							
$4I\,\frac{15}{2}$	2 MU	1	3	1	3	3	3	1	1
(108)	TH.	0	72	82	109	194	233	261	269
$4I\,\frac{13}{2}$	2 MU	3	1	3	3	1	3	1	
(6600)	TH.	6517	6618	6620	6637	6693	6704	6719	
$4I\,\frac{11}{2}$	2 MU	3	1	1	3	1	3		
(10250)	TH.	10221	10290	10296	10306	10325	10334		
$4I\,\frac{9}{2}$	2 MU	1	1	3	1	3			
(12400)	TH.	12366	12424	12445	12451	12538			
$4F\,\frac{9}{2}$	2 MU	1	3	3	1	1			
(15250)	TH.	15217	15251	15314	15319	15378			
$4S\,\frac{3}{2}$	2 MU	1	3						
(18350)	TH.	18346	18423						
$2H\,\frac{11}{2}$	2 MU	1	1	3	3	1	3		
(19150)	TH.	19143	19163	19190	19209	19244	19250		
$4F\,\frac{7}{2}$	2 MU	1	3	3	1				
(20450)	TH.	20444	20476	20481	20588				
$4F\,\frac{5}{2}$	2 MU	3	1	3					
(22100)	TH.	22127	22127	22179					
$4F\,\frac{3}{2}$	2 MU	1	3						
(22500)	TH.	22518	22599						
$2G\,\frac{9}{2}$	2 MU	1	3	1	1	3			
(24550)	TH.	24518	24586	24594	24608	24670			
$4G\,\frac{11}{2}$	2 MU	1	1	3	3	1	3		
(26400)	TH.	26379	26389	26419	26474	26505	26547		

TABLE 17. ENERGY LEVELS FOR Tm^{3+} IN KY_3F_{10}

LSJ state (centroid)		Energy level							
3H 6	2 MU	4	4	2	0	4	2	4	2
(170)	TH.	0	0	333	353	389	409	420	439
	2 MU	0	0						
	TH.	446	465						
3F 4	2 MU	0	2	0	0	4	2	4	
(5900)	TH.	5940	6006	6015	6016	6101	6191	6210	
3H 5	2 MU	2	0	0	2	4	0	4	2
(8400)	TH.	8309	8568	8603	8610	8628	8643	8669	8676
3H 4	2 MU	0	0	2	4	4	0	2	
(12700)	TH.	12708	12726	12882	12896	12909	12912	12956	
3F 3	2 MU	0	2	4	4	2			
(14500)	TH.	14615	14670	14674	14701	14722			
3F 2	2 MU	2	0	4	4				
(15100)	TH.	15217	15283	15340	15378				
1G 4	2 MU	0	0	0	2	4	2	4	
(21350)	TH.	21337	21338	21378	21474	21601	21678	21753	
1D 2	2 MU	0	2	4	4				
(28000)	TH.	28078	28147	28252	28266				
1I 6	2 MU	2	0	0	4	4	2	4	4
(34900)	TH.	34734	34851	34858	34893	34893	35063	35234	35328
	2 MU	2	0						
	TH.	35436	35486						
3P 0	2 MU	0							
(35500)	TH.	35677							
3P 1	2 MU	2	0						
(36400)	TH.	36518	36697						
3P 2	2 MU	2	0	4	4				
(38250)	TH.	38285	38441	38534	38628				
1S 0	2 MU	0							
(79590)	TH.	79769							

The phenomenological B_{kq} for Pr^{3+} in KY_3F_{10} and the odd-k A_{kq} given in table 2 (for an F charge of $q = -1$) were used along with the radial integrals given elsewhere[6] to calculate electric and magnetic dipole transition probabilities between the Stark split energy levels for Pr^{3+}. Branching ratios and lifetimes also were determined. The salient features of these calculations are given in tables 19 and 20. In table 19, the branching ratios from the fluorescing $^3P_0$ multiplet to the

[6] Clyde A. Morrison, Nick Karayianis, and Donald E. Wortman, Rare-Earth Ion-Host Lattice Interactions 4. Predicting Spectra and Intensities of Lanthanides in Crystals, Harry Diamond Laboratories HDL-TR-1816 (June 1977).

TABLE 18. ENERGY LEVELS FOR Yb^{3+} IN KY_3F_{10}

LSJ state (centroid)		Energy level			
2F $\frac{7}{2}$	2 MU	3	1	3	1
(250)	TH.	0	126	299	490
2F $\frac{5}{2}$	2 MU	3	3	1	
(10450)	TH.	10266	10361	10664	

TABLE 19. BRANCHING RATIOS FROM $^3P_0$ ENERGY LEVEL TO LOWER LYING J-MULTIPLETS FOR Pr^{3+} IN KY_3F_{10} AND IN YLF

Branching ratio	$^3P_0$ to (J-multiplet)	KY_3F_{10}	YLF
β_1	($^3H_4$)	0.33	0.36
β_2	($^3H_5$)	0.042	0.041
β_3	($^3H_6$)	0.32	0.50
β_4	($^3F_2$)	0.19	0.021
β_5	($^3F_3$)	0.0053	0.0056
β_6	($^3F_4$)	0.057	0.057
β_7	($^1G_4$)	0.053	0.022
β_8	($^1D_2$)	0.00033	0.00003

TABLE 20. ELECTRIC DIPOLE TRANSITION PROBABILITIES FROM FLUORESCING $^3P_0$ ENERGY LEVEL TO $^3H_4$ ENERGY LEVELS FOR Pr^{3+} IN KY_3F_{10} AND IN YLF

KY_3F_{10} [a]				YLF [b]			
1 [c]	2	3	4	1 [c]	2	3	4
2.168×10^{-5}	σ	E	0	9.674×10^{-6}	π	Γ_2	0
0	—	B_1 or B_2	70 [d]	1.600×10^{-5}	σ	$\Gamma_{3,4}$	79
3.813×10^{-6}	π	A_1 or A_2	166	—	—	Γ_1	217 [d]
0	—	B_1 or B_2	175	—	—	Γ_1	220
5.851×10^{-6}	σ	E	217	6.183×10^{-8}	σ	$\Gamma_{3,4}$	496
1.585×10^{-12}	π	A_1 or A_2	510 [d]	—	—	Γ_1	512 [d]
1.196×10^{-5}	π	A_1 or A_2	540 [d]	8.458×10^{-7}	π	Γ_2	514 [d]

[a] The $^3P_0$ is denoted A_1 and is at 20,728 cm^{-1} in KY_3F_{10}.
[b] The $^3P_0$ is denoted Γ_1 and is at 20,866 cm^{-1} in YLF.
[c] The most intense $^3P_0$ to $^3H_4$ transition was measured to go to the ground state for Pr^{3+} in KY_3F_{10} and in YLF. Measurements show that the next most intense transitions go to a 79-cm^{-1} level in YLF and to the 166- and 217-cm^{-1} levels in KY_3F_{10}.
[d] Calculated value.

Notes:

1: Electric dipole transition probability from $^3P_0$.
2: Polarization of emitted radiation.
3: Final state group property designation; the point symmetry at the impurity ion site is C_{4v} in KY_3F_{10} and is S_4 in YLF.
4: Final state energy position in units cm^{-1}.

lower lying J-multiplets are given. Corresponding quantities for Pr^{3+} in YLF are shown for comparison. Similar values of about 0.33 are obtained for β_1 for both materials, which is the first indication that $Pr^{3+}:KY_3F_{10}$ might be as good a blue-green laser as Pr^{3+}:YLF would be. In table 20, the $^3H_4$ energy levels and the individual line-to-line electric dipole transition probabilities are given. These calculations show that the largest transitions from the $^3P_0$ level in either material would be to the lowest lying energy levels. However, for Pr^{3+}:YLF, the higher of the possible laser terminal levels is at 79 cm^{-1}; this occurrence suggests that even at 77 K, Pr^{3+}:YLF acts as a three-level system. For Pr^{3+} in KY_3F_{10}, the higher of the possible laser terminal levels is experimentally determined to be at 217 cm^{-1}, and this occurrence might allow four-level laser operation at 77 K. (The Boltzman factor, $e^{-\Delta E/kT}$, which partially determines the population of the terminal level, is 0.14 for the 217 cm^{-1} KY_3F_{10} Pr^{3+} level and is 0.49 for the 79 cm^{-1} YLF level.) On the basis of the above discussions and the results obtained in the above calculations, further work to determine the feasibility of $Pr^{3+}:KY_3F_{10}$ as a blue-green laser seems warranted at this time.

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